Effect of Lead Extraction by EDTA on the Physico-Mechanical Behavior of Bentonite Soil

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Abstract: Lead contamination of soils is a common problem throughout the world. The application of ethylenediaminetetraacetic acid (EDTA) as an extraction material has been exhibited to be an effective method of removal of Pb from many contaminated soils. The extraction of heavy metal from contaminated soils will alter the properties of soil-water system, resulting in changes in the physico-mechanical behavior of soil, specifically soils with relatively high surface area such as bentonite. This study investigates the influence of lead removal from contaminated bentonite by the use of EDTA on its physic-mechanical behaviour. A set of experiment including precipitation testing, Atterberg limits, and consolidation were performed on contaminated bentonite before and after EDTA treatment. The achieved results were compared with bentonite behaviour. The results of this research indicate that in contaminated bentonite with medium concentration of lead nitrate (up to 10 cmol/kg soil) after soil remediation with EDTA, the mechanical behavior of soil is changed. After contamination removal, this mechanical behavior is relatively similar to the mechanical behavior of uncontaminated soil. In other words, the soil properties are almost reversible. However, in highly contaminated bentonite (i.e. 50 to 100 cmol/kg soil lead nitrate), there is not much difference between the mechanical properties of contaminated and remediated soil. In other words, in this case the soil properties after contaminant removal are mainly irreversible.

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1. Introduction

As a result of increasing world population and development of the agricultural and industrial fields in most of the countries, significant parts of the soils have been contaminated by organic and inorganic contaminants (Jones, 1991). Heavy metals (HMs) are the most common inorganic contaminants in urban areas and mines industrial wastewater (Kim et al., 2003; McIntyre, 2003). Among the heavy metals, lead (Pb) is the most abundant and most toxic contaminant (Yong, 2001). During the last decades significant progress and developments have made in the remediation of soils. One of the most effective methods for remediation of contaminated soils that contains lead (Pb) is soil washing and extracting the lead (Pb) from the soil by chelating agents. Chelating agents, by creating stable complexes with heavy metals, change the phases of these contaminants into liquid and extract them from the soil. Chelating agents have the least effect on the physical-chemical and biological properties of the soil (Lim et al., 2004). Based on previous research, Ethylene Diamnie Triacetic Acid (EDTA) is one of the most common and strongest chelating agents and it is used for extracting the lead (Pb) from contaminated soils (Lestan et al., 2008; Zhang et al., 2008). Because of high Cation Exchange Capacity (CEC) existing clay minerals in the soil are able to interact with heavy metal contaminants in the industrial wastewater and adsorb them by different mechanisms (Harter, 1983). Cation Exchange Capacity (CEC) is a criterion for determining the soil's ability for cation exchange and saving it. Because bentonite contains a high amount of montmorillonite and CEC and negative charge it is applied in environmental geotechnical projects (such as soil barriers in the landfill centers), liner systems, Geosynthetic Clay Liner (GCL) and clay liner for burying radioactive wastes (Pusch, 1994). As a result of the alkaline property of most of clay soils and low solubility rate of lead (Pb) in alkaline ambient, chelating solutions such as EDTA are used instead of pure water for washing contaminated soils that contain lead (Pb) (Lestan et al. 2008; Peters, 1999). As observed in previous studies, the mechanical properties of clay soils such as shear strength, consolidation, and permeability are strongly influenced by changes in pore water characteristics (concentration and capacity of cations, pH of system) and changes in their intermolecular forces (Mesri & Olson, 1971; Ouhadi et al., 2006; Yong 1992). Also experiments that are performed on the bentonite soil in this study have shown adsorbing heavy metal contaminants such as lead (Pb) and zinc (zn) change the pore water characteristics and intermolecular forces and as a result, mechanical and engineering properties of soil will be changed (Ouhadi et al., 2006).

As mentioned above, it seems inevitable to consider the extraction of heavy metal contaminants from soil and its effects on the pore water characteristics and finally changes in mechanical and engineering properties of clay soil. Various studies have been conducted during the last few years about remediation of heavy metals by using EDTA. These studies contain issues such as impacts of various factors on efficiency of remediation (pH, EDTA, concentration of contaminants) and reuse of washing solution. A lack of data about changes in mechanical properties of soil after remediation of contaminants by using chelating agents can be seen. In this present study, impacts of remediation of lead (Pb) contaminants by EDTA chelating agents on the mechanical properties of clay bentonite soil have been considered.

2. Material and Methods

2.1. Bentonite

Some physical and geo-environmental properties of the bentonite used in this study are shown in Table 1. The commercial name of applied bentonite for this research is Plateau of Iran Bentonite, and it is provided by Iran Barit Company.

Table 1. Physical and geo-environmental peoperties of bentonite (Ouhadi et al., 2006)

Parameter	**Data**
Soil Classification	CH
LL (%)	314.5
PI (%)	283.3
CEC(cmol/kg-soil)	68.2
Gs	2.79
pH	9.5
SSA(cm^2/g)	4130
Clay	76%
Silt	23%
Sand	1%
Water content (air-dried)	5.9
Water content (oven-dried)	7.1

2.2 Ethylene Diamine Triacetic Acid (EDTA)

EDTA sodium salt (Na_2EDTA), chemical formula of $C_{10}H_{14}N_2Na_2O_8.2H_2O$, has been used for this study.

2.3 Lead (Pb)

For simulating the contamination of the soil, $Pb(NO_3)_2$ solution was applied by various concentrations.

2.4 Preparation of contaminated samples

For consideration of interaction between lead (Pb) and bentonite and determining the rate of adsorbing and retention lead (Pb) in the sample soil, and also preparing contaminated samples, at the beginning, the proper amount of dry soil (2 g for environmental tests and 100 g for mechanical tests) was weighted with accuracy of 0.001 g and was accumulated in suitable containers (50 ml centrifugal tubes or 1.5 liter plastic bottles) and then lead nitrate contamination solution (1-250 cmol/kg-soil) was added to soil. The ratio of suspension solution and soil suspension must always be constant, 1:10. It helps to compare all the tests with each other (environmental and mechanical). Samples were kept at room temperature for 96 hours. These samples were put in the horizontal shaker for two hours every day during this time to satisfy the equilibration and interaction conditions. After 96 hours, samples that were in the 150 ml tubes were centrifuged by 4000 rpm for 15 minutes and liquid and solid phases were separated and then analyzed using an Atomic Adsorption Spectrophotometer (AAS). By subtracting the amount of remaining lead (Pb) in the liquid phase from the amount of lead (Pb) in the contamination solution, the amounts of retained lead (Pb) in the sample contaminated soil were calculated.

2.5 Method of remediation of contaminants and preparing the samples

After preparing the contaminated bentonite samples, those with suitable concentration (0.01-0.1 molar) were selected and were exposed to various concentrations of EDTA for remediation. All the contaminated samples (both inside the tubes and inside the bottles) were dried in the oven at 35 degree Celsius and then washed using a washing solution with determined concentration and also a constant ratio of 1:10 for washing solution – contaminated soil. To find the optimal amount of EDTA for full remediation of lead (Pb) from the soil, each sample was washed with EDTA with various concentrations (1, 5, 10, 50 and 70 cmol/kg-soil). The equilibration step and washing the contaminated soil is similar to what was explained in the previous section (preparing contaminated samples). The only difference was the use of EDTA solution as lead (Pb) nitrate solution and the use of contaminated dried soil as natural soil. Finally, extracted solution from the samples inside the centrifuge tubes was analyzed using an Atomic Adsorption Spectrophotometer (AAS). The samples inside the 1.5 liter bottles were poured inside the Teflon tubes with internal diameter of 7 cm and height of about 15 cm for mechanical tests. Also Teflon pistons were fabricated with longitudinal slots for draining and an external diameter of less than 7 cm to do the easier loading for extracting the additional water and preparing the initial samples for mechanical tests (liquid limit, and consolidation tests). After the pre-consolidation process, an undisturbed sample was prepared from samples inside the Teflon tubes and the rest of sample was used for liquid limit test. For sediment testing, 500 ml of suspension from inside the 1.5 liter bottles was poured in the graduated cylinder with a capacity of 1000 ml and then the volume of suspension inside it was increased by adding distilled water up to 1000 ml. Then, samples inside the graduate cylinder were located on a smooth surface for 24 hours to observe the sedimentation rate.

3. Results and Discussions

3.1 Environmental tests

3.1.1 Interaction between bentonite and lead (Pb) contaminant

According to the results that are shown in Fig.1, for concentrations around 0.07 molar (70 cmol/kg-soil) that are equivalent to the concentration of CEC of soil, almost all of the heavy metal ions inside the electrolyte remained in the soil. Up to this concentration, the graph has a linear behavior and the slope of the graph is equal to 45 degree and it means the amount of lead (Pb) inside the initial solution and amount of adsorbed lead (Pb) by the soil are equal. By increasing the concentration of contamination to more than CEC, the bentonite sample is not able to retain all the ions of the heavy metal, lead (Pb), and adsorption trend (slope of the graph) decreases drastically. According to constant level of adsorption of lead (Pb) in the soil for concentrations of more than 0.07 molar (70 cmol/kg-soil), an interval of concentrations from 0.001 molar up to 0.01 molar (1-100 cmol/kg-soil) was chosen to prepare the contaminated samples in this study.

3.1.2 Extracting lead (Pb) from contaminated bentonite by EDTA and determining the optimum amount of lead (Pb)

The amount of extracted lead (Pb) that is obtained from washing contaminated soil by distilled water is presented in Figure 2. For the entire tests, the constant ration of 1:10 (solution: soil) is applied. As can be seen, the amount of extraction of lead (Pb) goes up by increasing the concentration of EDTA. Also, it is obvious that the EDTA solution in each specific concentration has extracted a specific amount of lead (Pb) from soil that is equal to its own concentration. In fact, the minimum required concentration for EDTA (optimum concentration) for perfect extraction of the lead (Pb) contaminant from the contaminated soil is exactly equivalent to the existing contaminant in the soil. The “equivalent or equal concentration” is defined as the condition in which the EDTA concentration is equal to the contaminant concentration in soil. For example, the equivalent concentration for 30 cmol/kg-soil means that the bentonite soil contains 30 cmol/kg-soil lead (Pb) contaminant and this contaminated soil is washed by EDTA solution with a concentration equal to 30 cmol/kg-soil and the whole amount of lead (Pb) inside the soil is extracted and the soil contains no more contaminant.

3.2 Behavioral and mechanical tests

3.2.1 Sedimentation test

For observing effects of lead (Pb) contaminant on structures of the bentonite, sedimentation tests for bentonite samples were performed in various concentrations of lead (Pb). Twenty-hours hours after pouring the suspension samples inside the similar graduated cylinders, some photos were taken and the photos are shown in Fig 3. According to Fig 3, by increasing the concentration of lead (Pb) and replacing it as alkaline metals in the soil, repulsive forces between the particles and the negative charges of the particles are decreased. As a result of this phenomenon, a more concentrated and flocculated double layer is created. By mounting the concentration of the contaminant, flocculating of the system increases sharply and the solid phase of soil get separated from the liquid phase rapidly. Levels of suspension sedimentation after complete remediation of contaminants by an equivalent concentration of EDTA are shown in Fig 4. Despite the complete remediation of the lead (Pb) from the bentonite for all concentrations, significant differences can be seen. This is because of the effects of exchangeable cations on the soil. By increasing the equivalent concentration, the amount of existing lead (Pb) in the soil was mounted. Based on previous studies, sodium montmorillonite satisfies the double-layer theory in comparison with calcium montmorillonite and it also acts dependent to double-layer and intermolecular forces (Mitchell, 1993). Here, by increasing the concentration of the lead (Pb) in the soil, the amount of sodium is decreased and its impressibility of double-layer thickness and intermolecular forces decreases. Extracted results from this study have reasonable similarities with previous research (Mesri & Olson, 1971).

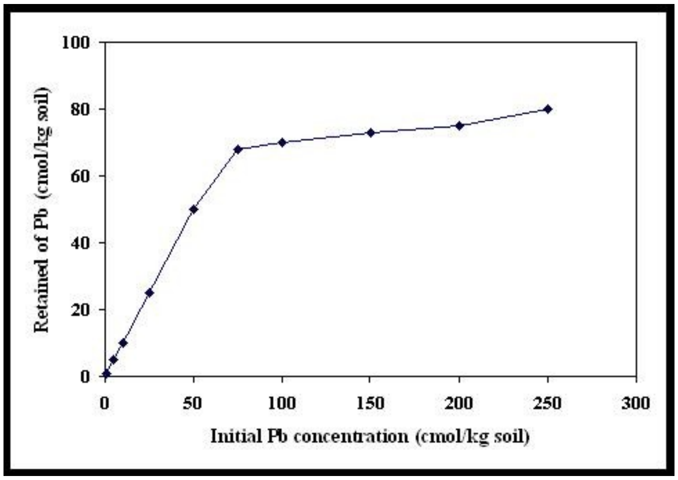

Fig 1. Specifications of adsorption and retention of lead (Pb) in the bentonite

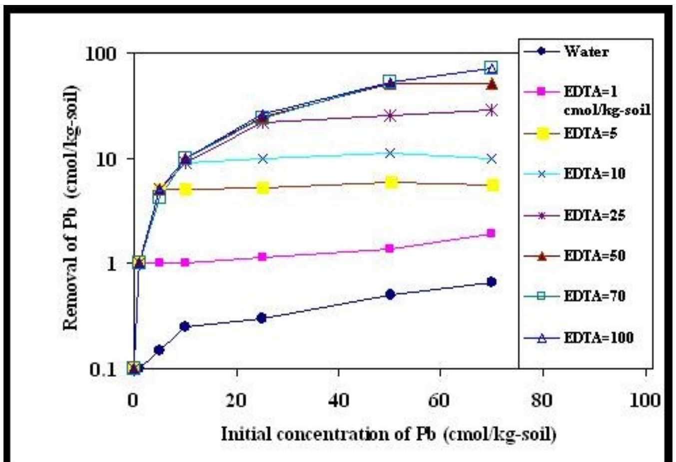

Fig 2. Amounts of extracted lead (Pb) by various concentrations of EDTA

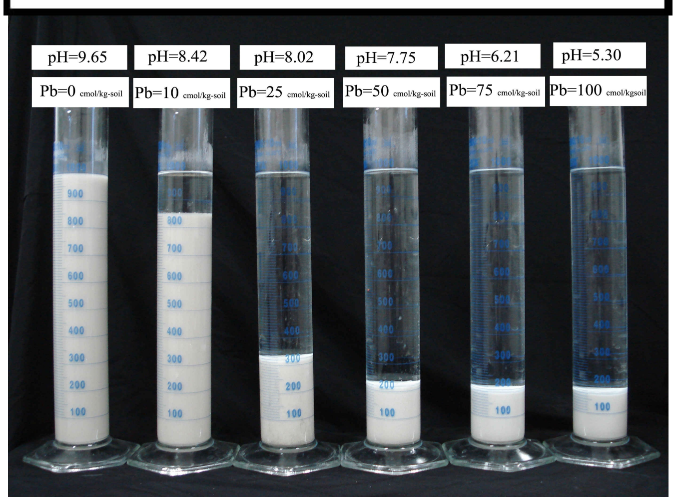

Fig 3. Effects of lead adsorption on sedimentation levels of bentonite for various concentrations after 24 hours

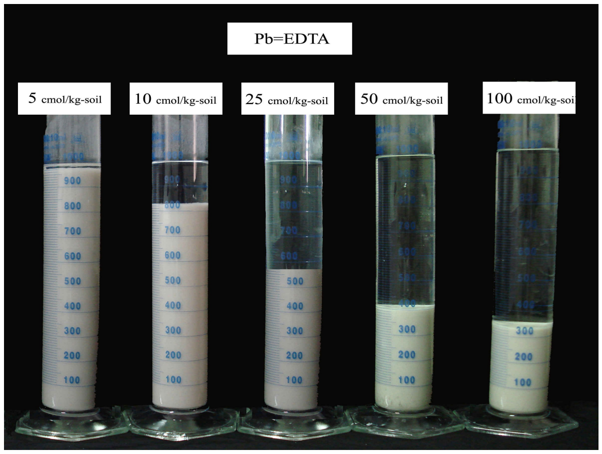

Fig 4. Comparison between sedimentation levels of bentonite for various equivalent concentrations after 24 hours

3.2.2 Liquid limit test

Effects of the various concentrations of the lead (Pb) contaminant on the liquid limit of bentonite are shown in Fig 5. As indicated, by mounting the concentration of lead (Pb) contaminant, a significant downward trend appears in the liquid limit of contaminated soil. Levels of extracted liquid limit from the test performed on the bentonite with complete remediation of lead (Pb) are illustrated in Fig 6. By comparing Fig 5 and 6 it is clear that that after remediation of lead (Pb), only in low concentrations (5 and 10 cmol/kg-soil) significant changes near to liquid limit appear and after complete remediation of the lead (Pb) liquid limit of contaminated soil approaches the liquid limit of natural bentonite. In high concentrations (50 and 100 cmol/kg-soil) even after complete remediation of lead (Pb) from the soil there are no significant changes in liquid limit of the soil. Increasing initial liquid limit in concentration of 5 cmol/kg-soil is related to fully saturating the montmorillonite with sodium and different rheological behavior in low concentrations of sodium (van Olphen, 1977; Sivapullaiah, 2000). By decreasing the amount of existing sodium in the soil and dissimilar behavior of the soil with the double-layer theory and intermolecular forces, it is clear that irreversibility of the bentonite characteristics to the natural conditions reduces drastically.

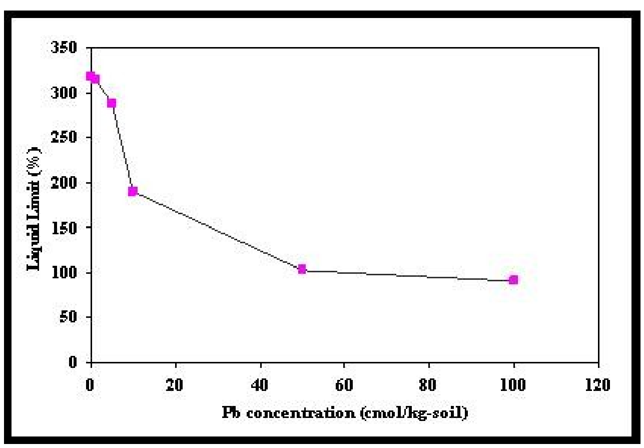

Fig 5. Effect of various concentrations of lead (Pb) on liquid limit of bentonite

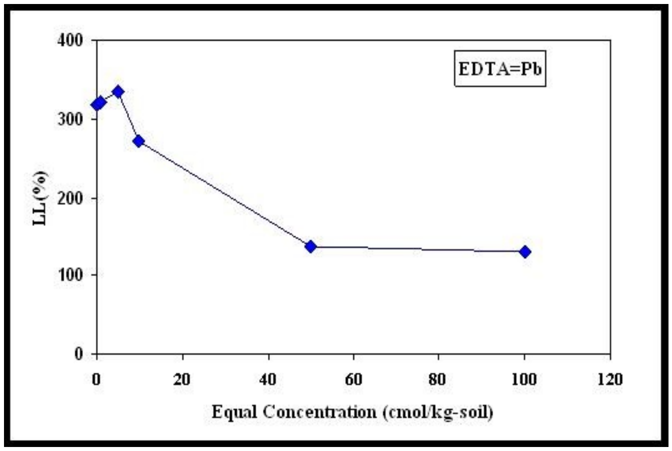

Fig 6. Liquid limit of bentonite after full remediation by EDTA in various equivalent concentrations

The reason for little differences between equivalent concentrations of 50 and 100 cmol/kg-soil could be the clay minerals that are saturated by lead (Pb) and bivalent alkaline cations in equivalent concentration of 50 cmol/kg-soil. In the soil that contains 50 cmol/kg-soil of lead (Pb) in addition to 50cmol/kg-soil lead (Pb) inside the soil, there are also 20cmol/kg-soil bivalent alkaline cations and it fills the rest of soil adsorption capacity. The statistics revealed no significant differences between the two soil samples.

3.2.3 One-dimensional consolidation test

The graphs related to the void ratio against the consolidation pressure for various concentrations of lead (Pb) contaminant in the contaminated soil are illustrated in Fig 7. Similar to what earlier results, by increasing the concentration of lead (Pb) contaminant and replacing it with existing sodium and calcium in the soil there was a significant decrease in the initial void ratio of the soil and consolidation properties of the contaminated soil.

Graphs related to void ratio in respect to the consolidation pressure in the bentonite after remediation of the contaminants by various concentrations of EDTA are illustrated in Fig 8. By comparing Fig 7 and 8 and regarding the complete remediation of lead (Pb) contaminant from all the samples, by increasing the equivalent concentration, a significant descending trend for the ratio of pores of the soil can be seen. With respect to the previous theory, because of the existence of lead (Pb) contaminant in the bentonite, intermolecular forces do not affect the bentonite significantly.

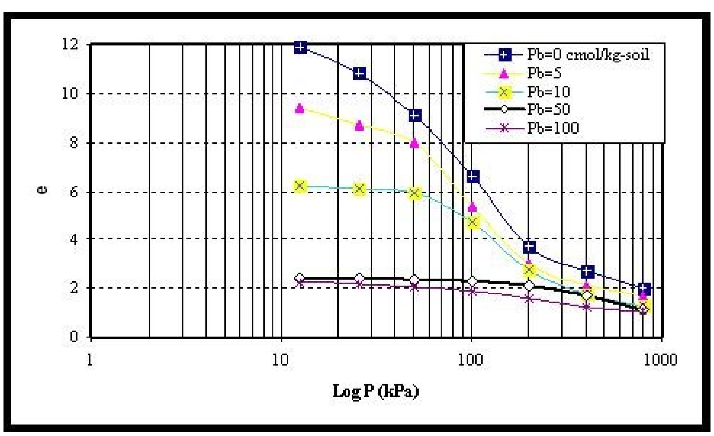

Fig 7. Void ratio via consolidation pressure for contaminated bentonite in various concentrations of lead (Pb)

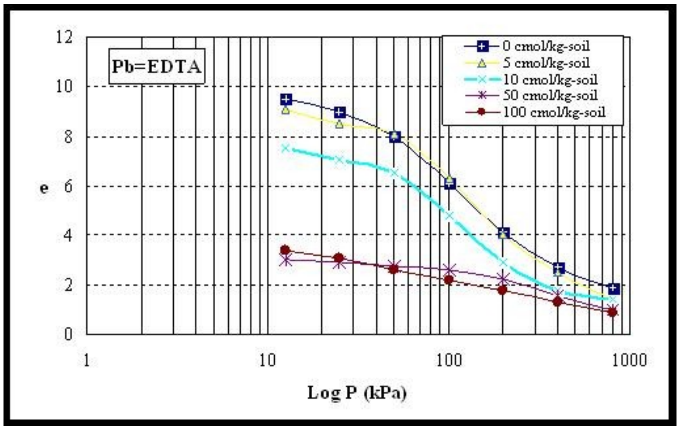

Fig 8. Void ratio via consolidation pressure for bentonite in various equivalent concentrations

Also, it can be seen that only in the low concentrations (5 and 10 cmol/kg-soil) EDTA was able to remediate the soil by extracting the lead (Pb) from the contaminated soil and made the properties of the contaminated soil similar to the natural bentonite by replacing lead (Pb) with sodium. Mounting the concentration of contaminant, properties of soil before and after remediation of soil do not change significantly and it has a perfect corresponding to the results of previous tests.

4. Conclusion

Effects of remediation of lead (Pb) contaminant by EDTA chelating agent on the behavioral and mechanical properties of bentonite soil were considered in this study. Based on the experiments of this study these results were extracted:

1. EDTA chelating agent is able to extract lead (Pb) contaminant from the contaminated bentonite exactly equal to its own concentration.

2. The optimum amount of EDTA for remediation of lead (Pb) from the bentonite soil is exactly equal to concentration of existing contaminant in the soil.

3. Mechanical properties and behavior of the contaminated bentonite (with low concentrations of lead (Pb), around 10 cmol/kg-soil) have changed after remediation by EDTA and have approached to the natural bentonite.

4. Mechanical properties and behavior of the contaminated bentonite (with high concentrations of lead (Pb), between 50- 100 cmol/kg-soil) have not changed significantly after remediation by EDTA and irreversibility in the properties of the soil have not been seen.

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